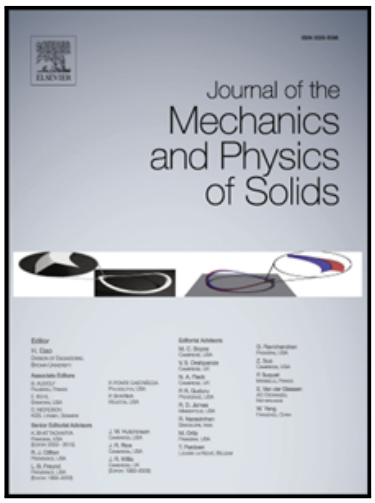

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PII: S0022-5096(17)30880-3
DOI: 10.1016/j.jmps.2018.03.008
Reference: MPS 3301

To appear in: *Journal of the Mechanics and Physics of Solids*

Received date: 30 September 2017
Revised date: 7 March 2018
Accepted date: 8 March 2018

Please cite this article as: J.P. Méndez, M. Ponga, M. Ortiz, Diffusive molecular dynamics simulations of lithiation of silicon nanopillars, *Journal of the Mechanics and Physics of Solids* (2018), doi: 10.1016/j.jmps.2018.03.008

Diffusive molecular dynamics simulations of lithiation of silicon nanopillars

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Abstract

We report diffusive molecular dynamics simulations concerned with the lithiation of Si nano-pillars, i. e., nano-sized Si rods held at both ends by rigid supports. The duration of the lithiation process is of the order of miliseconds, well outside the range of molecular dynamics but readily accessible to diffusive molecular dynamics. The simulations predict an alloy $Li_{15}Si_4$ at the fully lithiated phase, exceedingly large and transient volume increments up to 300% due to the weakening of Si-Si iterations, a crystalline-to-amorphous-to-lithiation phase transition governed by interface kinetics, high misfit strains and residual stresses resulting in surface cracks and severe structural degradation in the form of extensive porosity, among other effects.

Keywords: Li-based batteries, Li diffusion, Lithiation, Non-equilibrium statistical mechanics, Long-term diffusion simulations, Diffusive molecular dynamics.

1. Introduction

Si nanowires have been investigated as building blocks for anode applications in high energy-density Li batteries, reaching theoretical specific capacities of up to 4212 mAh/g (Aifantis et al., 2010). However, Si nanowires are hampered by the mechanical degradation that occurs during lithiation, a fundamental process that takes place when Li cations enter the Si lattice

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